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(12) **Patent:**

(11) **CA 601124**

(54) CONTINUOUS FLOW PROCESS FOR THE MANUFACTURE OF HYDRATED CALCIUM SILICATES

(54)

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ABSTRACT:

CLAIMS: [Show all claims](#)

\*\*\* Note: Data on abstracts and claims is shown in the official language in which it was submitted.

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(45) Issued: **July 5, 1960**

(22) Filed:

(43) Laid Open:

(52) Canadian Class (CPC): **23/190**

(51) International Class (IPC): **N/A**

Patent Cooperation Treaty (PCT): **No**

(30) Application priority data: **None**

Availability of licence:

N/A

Language of filing:

Unknown

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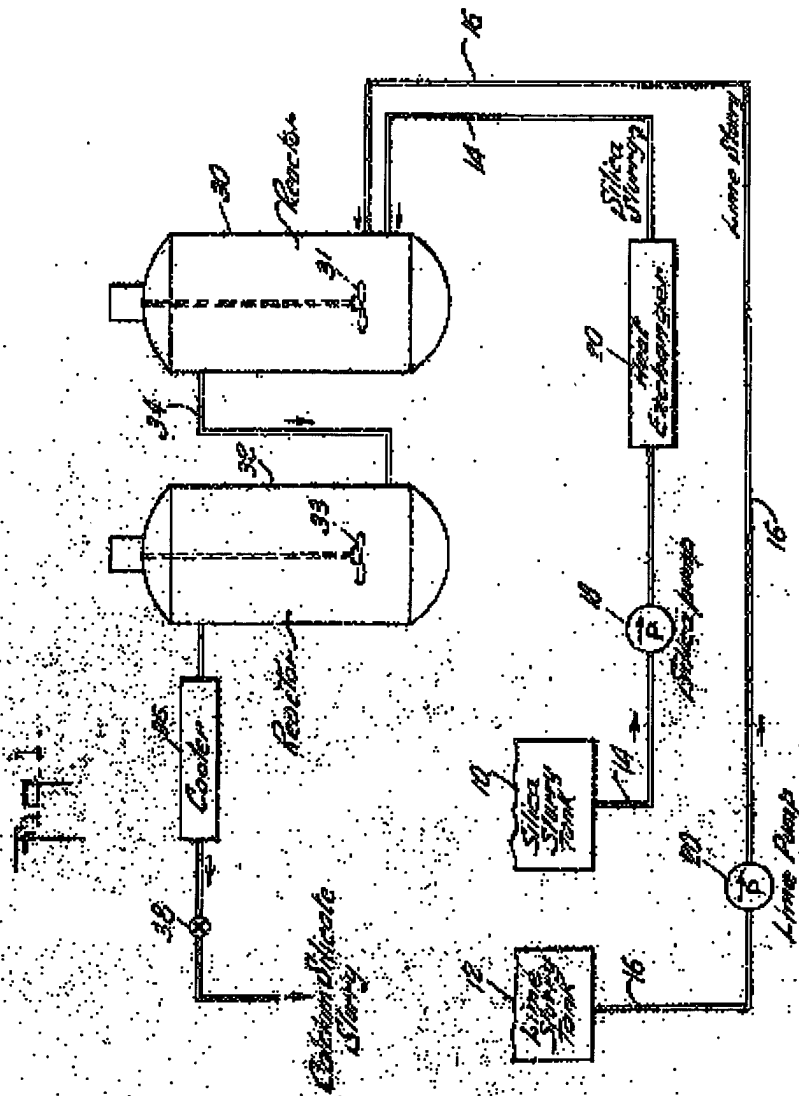
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CANADA

00601124



INVENTOR

Charles R. Knaack

PATENT AGENT

Virgil C. Kline

Fig. 2.

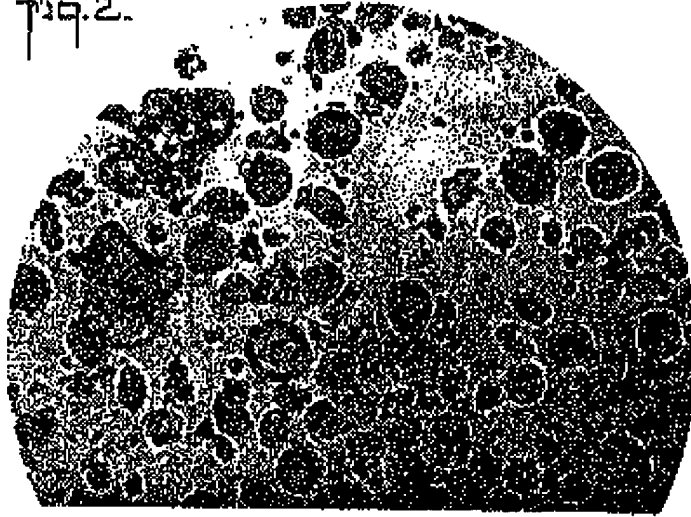


Fig. 3.



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10/25/94

Fig. 4.

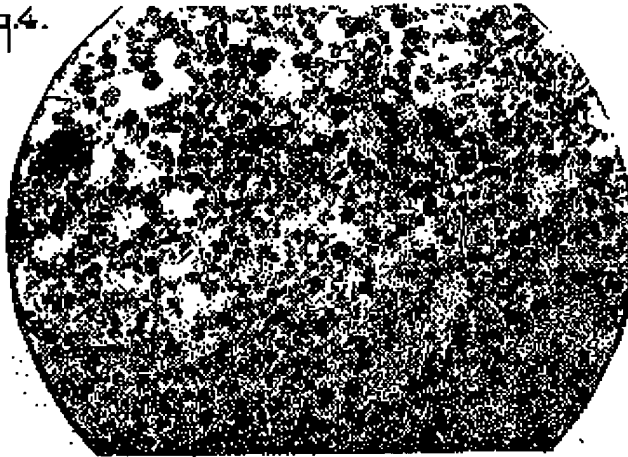
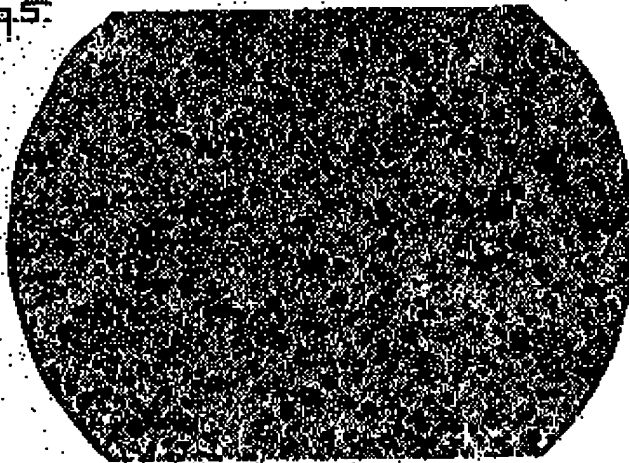


Fig. 5.



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CROSS

FIG. 6

Fig. 6.

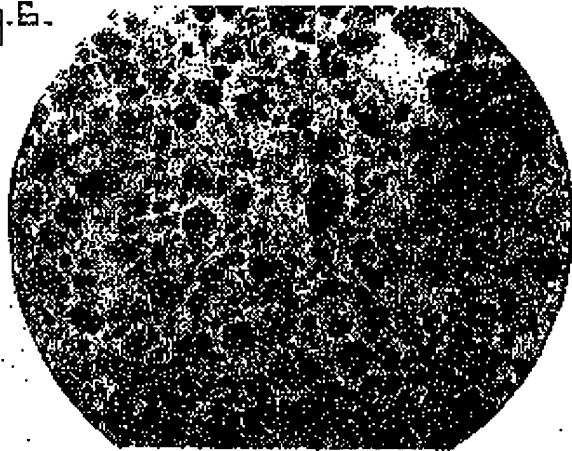
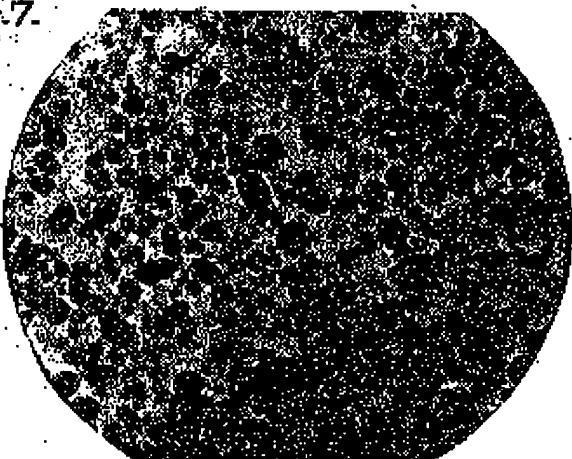


Fig. 7.



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1 This invention relates to the manufacture of hydrated  
2 calcium silicates, and more particularly to the manufacture  
3 of hydrated calcium silicates by a continuous flow process.

4 The preparation of hydrated calcium silicate by the  
5 hydrothermal reaction of lime and the finely divided reactive  
6 silica has been heretofore proposed. For example, United  
7 States Letters Patent No. 1,574,363, to Calvert exemplifies  
8 a typical hydrothermal manufacturing procedure consisting of  
9 a batch-type reaction including the steps of charging a  
10 reaction vessel with a slurry of finely divided lime and  
11 siliceous material, such as diatomaceous silica, and then  
12 heating the material to reaction temperatures of the order of  
13 212°F. or higher if a pressure vessel is employed, for a time  
14 sufficient to effect completeness of the reaction. Typical  
15 products resulting from this type of reaction are a finely  
16 divided particulate or granular porous material having a  
17 fluffed bulk density 4 to 7 lbs. per cu. ft., with the bulk  
18 density decreasing with higher reaction temperatures. The  
19 wet cake density of such dried particulate hydrated calcium  
20 silicates varies from 9 to 11 lbs. per cu. ft. These products  
21 also have very high absorbent properties, e.g., their water  
22 absorption as measured by a Gardner-Coleman test normally  
23 consists of about 375% to as high as 700%, with absorptive  
24 capacities increasing with increased reaction temperatures.  
25 These highly absorbent, low bulk density calcium silicates  
26 are useful as a carrier for insecticides or other liquid  
27 chemicals and as a bulking agent for dry powder formulations,  
28 among other applications.

29 Synthetic hydrated calcium silicates formed from the  
30 batch process, however, have not been found suitable for some  
31 uses where it is required that the product be (1) of such  
32 structure and size that it is a very free-flowing powder, and  
33 (2) of heavier bulk density than the batch process product

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1 and yet have good absorbent characteristics. Hydrated cal-  
2 cium silicate powders produced by a batch process have not  
3 been suitable for use as a carrier in some insecticide formu-  
4 lations due to their bulk. This is particularly true in high  
5 concentrate wettable powders prepared for overseas shipment  
6 where freight rates are based on cubage.

7 Another application for an absorbent material is a filler  
8 for containers in which are stored, normally under pressure,  
9 liquid petroleum products, such as low-boiling hydrocarbons.  
10 It is apparent that for such service suitable fillers must  
11 have a high bulk density and a good degree of absorbency per  
12 unit volume of material, to enable substantial amounts of  
13 absorbent and, hence, of liquid to be packaged in the contain-  
14 er. Also, the absorbent should have a structure which makes  
15 it freely flowable in order that the container may be easily  
16 filled. Hydrated calcium silicates prepared by the batch  
17 reaction product as described by Calvert have not been fully  
18 suitable as a filler for such an application due to their  
19 relatively low bulk density and inadequate flowability.

20 It is a principal object of this invention to provide an  
21 improved hydrothermal method of manufacturing discrete hydra-  
22 ted calcium silicates of increased bulk density.

23 It is a further object of this invention to provide a  
24 method of hydrothermally preparing particulate hydrated cal-  
25 cium silicates with a substantially uniform and large particle  
26 structure rendering them freely flowable and handleable.

27 It is a still further object of this invention to pro-  
28 vide a means of controlling or increasing bulk density, regu-  
29 lating the particle size and absorption capacity of hydro-  
30 thermally prepared, particulate hydrated calcium silicates.

31 These and other objects and advantages of the invention  
32 will become apparent when consideration is given to the here-  
33 inafter detailed description thereof, taken together with  
34 the accompanying drawings wherein:



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1 Fig. 1 is a schematic flow diagram of the preferred pro-  
2 cedure for manufacturing hydrated calcium silicates in  
3 accordance with this invention;

4 Figs. 3, 4 and 6 are photomicrographs (500X) of hydrated  
5 calcium silicates formed in a batch-type reaction; and

6 Figs. 2, 5 and 7 are photomicrographs (500X) of hydrated  
7 calcium silicates formed in accordance with this invention.

8 In the development of processes involving chemical  
9 reactions, it is normal practice to study the reaction in a  
10 small batch-type reactor. Data, such as the relation of  
11 extent of conversion, type of product, etc., to the reaction  
12 conditions such as time, temperature, pressure, etc., are  
13 usually obtained from these studies. Although it is entirely  
14 feasible to carry out these same reactions on a large scale  
15 using a batch reaction, it is often desirable to go to con-  
16 tinuous processes due to the need for less equipment for  
17 operation and generally lower manufacturing costs. The  
18 relationship between product characteristics, percentage con-  
19 version, etc., obtained by batch reaction and continuous  
20 reaction, in a series of vessels, has been described in the  
21 literature (e.g., MacMullin, R. B., and Weber, K., Jr., Chem.  
22 & Met. Engr. 52 No. 5, 101 (1945), and Weber, A.P., Chem.  
23 Engr. Progress, 49, 26 (1953)), and is a generally accepted  
24 chemical engineering principle. It is standard practice to  
25 design a continuous flow process from batch reaction data  
26 with the expectation of obtaining products with substantially  
27 identical characteristics if similar reaction conditions are  
28 used and a plurality of reactors are connected in series.  
29 The more reactors that are connected in series, the more  
30 nearly the percentage conversion approaches that obtained in  
31 a batch process. For example, with three reactors employed,  
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1 about 98 percent conversion can be expected; an infinite  
2 number of reactors would be necessary for 100 percent con-  
3 version.

4 This invention indicates that this known principle of  
5 chemical engineering does not apply in the manufacture of  
6 hydrothermal calcium silicates. It has been found that the  
7 hydrothermal reaction of lime and finely divided reactive  
8 silica in accordance with this invention results in a product  
9 having a high bulk density, a free-flowing rounded particle  
10 shape and structure, and yet relatively good absorbent  
11 characteristics.

12 The products of this invention are formed by a procedure  
13 wherein lime and silica are fed continuously to one or more  
14 reaction vessels in series, and the hydrated calcium silicate  
15 reaction product continuously removed at a rate equivalent  
16 to the rate of feed of the reactants. Preferably, at least  
17 two reactors in series are employed to insure complete  
18 reaction, but it is possible to obtain the product of this  
19 invention using only one reactor on a continuous basis.

20 Finely divided lime and finely divided reactive sili-  
21 ceous material, such as diatomaceous earth, quartz, etc., are  
22 employed as starting materials. Preferably, finely divided  
23 hydrated lime and diatomaceous silica are employed. The  
24 hydrated calcium silicate of this invention may be formed  
25 employing these starting materials in a  $\text{CaO/SiO}_2$  mol ratio  
26 within the range of 0.1 to 2.0. As the initial step of the  
27 process, the finely divided lime and reactive silica are  
28 suspended in at least sufficient water to form a pumpable  
29 slurry, and, preferably, the slurry in which the reaction is  
30 to take place is formed with a water-to-solids ratio of about  
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1 4 to 50 parts of water per part of solids. The lime and  
2 silica may be suspended together to form a common slurry, or  
3 can be suspended individually to form separate slurries  
4 thereof. In the formation of the slurry or slurries, as the  
5 case may be, at least sufficient water is employed to render  
6 the consistency of the slurry fluid enough to enable it to be  
7 pumped to the reaction vessel. Preferably, such slurry con-  
8 centrations will range between 1/2 to 2 lbs. of solids per  
9 gallon of water.

10 In accordance with this invention, the slurry of lime  
11 and reactive silica in the reaction vessel is heated to a  
12 temperature of at least approximately 212°F. in order to  
13 effect the desired reaction. When an autoclave is employed  
14 as the pressure vessel, the upper temperature for reaction  
15 is limited only by the pressure capable of being maintained  
16 in the autoclave and the critical point of water. Tempera-  
17 tures within the range of approximately 212°F. to 500°F. are  
18 normally employed. The  $\text{CaO/SiO}_2$  mol ratio, the reaction  
19 temperature, and the reaction time, all have an effect on  
20 the type of calcium silicate formed in the reaction. If the  
21 reaction temperature is below about 360°F. a compound commonly  
22 known as calcium silicate hydrate I (Taylor, J. Chem. Soc.  
23 170 (1953)) is formed over the entire range of the  $\text{CaO/SiO}_2$   
24 mol ratio (0.1 to 2.0). At temperatures of about 212°F. this  
25 reaction, on a continuous basis, has been found to be complete  
26 in approximately 20 minutes. If the reaction temperature  
27 is above 360°F. and the  $\text{CaO/SiO}_2$  mol ratio is 0.1 to 0.8,  
28 calcium silicate hydrate I will be formed first, and then  
29 will be partially or completely converted to CS-55, a low  
30 solubility hydrated calcium silicate, having the formula

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1 2CaO.38SiO<sub>2</sub> with approximately 2 mols of water and an X-ray  
2 diffraction pattern having very strong lines  $d = 3.12\text{\AA}$  and  
3  $d = 4.12\text{\AA}$  and a medium line at  $d = 8.34\text{\AA}$ . A substantially  
4 complete conversion to CS-55 is obtained in about 2 hours  
5 if a CaO/SiO<sub>2</sub> mol ratio of 0.5 to 0.6 is used and the re-  
6 action temperature is 450°F. If the CaO/SiO<sub>2</sub> mol ratio is  
7 0.8 to 1.5 and the reaction temperature is in excess of about  
8 360°F., calcium silicate hydrate I first forms, and then this  
9 is partially or completely transformed to Xonotlite. A  
10 substantially complete conversion to Xonotlite can be obtain-  
11 ed in about 2 hours if a CaO/SiO<sub>2</sub> mol ratio of 1.0 is used  
12 and the reaction temperature is 450°F. It has been found  
13 that reaction times longer than these necessary to complete  
14 the reaction have no detrimental effect upon the physical  
15 characteristics of the product, although it may result in  
16 converting calcium silicate hydrate I to CS-55, or to Xonot-  
17 lite.

#### 18 Example I

19 The following is a typical example of this invention.  
20 For clarity, reference is made in this example to the sche-  
21 matic flow sheet illustrated in Fig. 1. A slurry of silica  
22 was prepared by mixing 50 lb. of finely ground diatomaceous  
23 earth in 100 gallons of water in a suitable container 10. A  
24 slurry of hydrated lime was prepared by mixing 200 lb. of  
25 hydrated lime in 100 gallons of water in another suitable  
26 container 12. These two slurries were pumped separately into  
27 the first reactor 30 by means of suitable transport lines  
28 14 and 16, respectively, and pumps 18 and 20, respectively.  
29 The silica slurry was pumped at a rate of about 420 gpm and  
30 the lime slurry at a rate of about 70 gpm. This was calcul-  
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1 ated to give a  $\text{CaO}/\text{SiO}_2$  mol ratio of about 0.6. The silica  
2 slurry was heated to about 350°F. by passing it through a  
3 conventional heat exchanger 20. The lime slurry was added to  
4 the reactor 30 at room temperature. Both the silica and the  
5 lime slurry were pumped to the reactor system continuously.  
6 The reactor system consisted of two agitated autoclaves 30  
7 and 32 connected in series, by means of transport line 34,  
8 each having a capacity of approximately 450 gallons and  
9 equipped with suitable agitators 31 and 33, respectively.  
10 The effective retention time in the reactors was 1.7 hr. The  
11 reaction temperature therein was maintained at about 300°F.  
12 The reacted product was continuously discharged from the  
13 second reactor 32 through a heat exchanger cooler 36 and a  
14 pressure reducing valve 38. After filtering, drying, and  
15 grinding in a conventional manner, the product had the follow-  
16 ing properties:

17 Bulk Density 12.6 lb./cu. ft.  
18 Gardner-Coleman Water Absorption 270 lb./100 lbs. solids

19 The product was identified by X-ray diffraction to be calcium  
20 silicate hydrate I.

#### 21 Example II

22 The following is a typical example of this invention  
23 when the reaction conditions are such that CS-55 is formed.  
24 The feed slurry was prepared by mixing finely ground dia-  
25 tomaceous earth and hydrated lime using the following propor-  
26 tions:

27 510 pounds of diatomaceous earth,  
28 290 pounds of hydrated lime, and  
29 500 gallons of water.  
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1 The reaction system consisted of two agitated autoclaves  
2 each having a working capacity of 375 gallons. Feed slurry  
3 was continuously pumped into the first autoclave at a rate of  
4 53 gallons per hour. Steam was added to the first autoclave  
5 at a rate sufficient to keep the slurry in the first auto-  
6 clave at a temperature of 450°F. Reacted slurry was continu-  
7 ously removed from the second autoclave through a water-  
8 cooled heat exchanger and a pressure reducing valve at such a  
9 rate that the amount of fluid in the autoclaves remained con-  
10 stant. After filtering, drying and grinding in a conventional  
11 manner, the product had the following properties:

12 Bulk Density 20 lbs./cu. ft.

13 Gardner-Coleman Water Absorption 140 per cent

14 The product was identified by X-ray diffraction to be CS-55.

15 Example III

16 The following is a typical example of this invention  
17 when the reaction conditions are such that Xonotlite is  
18 formed. The feed slurry was prepared by mixing finely ground  
19 diatomaceous earth and hydrated lime using the following pro-  
20 portions:

21 180 pounds of diatomaceous earth,

22 222 pounds of hydrated lime, and

23 500 gallons of water.

24 The reaction system consisted of two agitated autoclaves  
25 each having a working capacity of 375 gallons. Feed slurry  
26 was continuously pumped into the first autoclave at a rate of  
27 53 gallons per hour. Steam was added to the first autoclave  
28 at a rate sufficient to keep the slurry in the first autoclave  
29 at 450°F. Reacted slurry was continuously removed from the  
30 second autoclave through a water-cooled heat exchanger and a  
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1 pressure reducing valve at such a rate that the amount of the  
2 fluid in the autoclaves remained constant. After filtering,  
3 drying and grinding in a conventional manner, the product  
4 had the following properties;

5 Bulk Density 11 lb./cu. ft.

6 Gardner-Coleman Water Absorption 300 per cent

7 The product was identified by X-ray diffraction to be Xenot-  
8 lite.

9 It is to be appreciated that the equipment and proced-  
10 ural steps outlined in Examples I, II, and III are exemplary  
11 and the invention described herein is not limited thereto.

12 It is also to be appreciated that the use of heat exchangers  
13 as indicated is not essential to the described process, but  
14 renders the preferred embodiment of the invention more econ-  
15 omical. The use of direct steam heating as described in  
16 Examples II and III can be used to give the desired results.  
17 Further, it is to be appreciated that any suitable means for  
18 maintaining the slurries undergoing reaction at reaction  
19 temperatures may be employed, and any suitable apparatus for  
20 transporting the materials to the reaction chamber and  
21 removing them therefrom may be used. So far as the reaction  
22 chambers themselves are concerned, any suitable vessel and  
23 suitable means of agitation, may be employed, and as indicated  
24 hereinabove, any suitable numbers thereof can be used.

25 As indicated above, the products of this invention may  
26 be formed at varying reaction temperatures and with varying  
27 mol ratios of lime and  $\text{SiO}_2$ . All of the reaction products of  
28 this invention, however, indicate a distinction from a batch-  
29 type reaction product formed under the same conditions.  
30 This is clearly illustrated in Table I which shows the  
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1 typical differences in the batch and continuous process  
2 reaction products formed with various reaction temperatures  
3 and mol ratios of lime and silica. In each case hydrated  
4 lime and finely divided diatomaceous earth were reacted  
5 using similar reaction conditions (i.e., temperature,  
6 reaction time, slurry concentration), except for the indicat-  
7 ed deviations.

8       The dry flowability of the powder was determined by  
9 measuring the time for a given volume of the dry powder to  
10 flow out of a vibrating funnel. Volume was used as a basis  
11 of measurement, rather than weight because of the wide vari-  
12 ation in bulk density of the products. If the measurements  
13 were made on a weight basis, the continuous flow process  
14 product would show an even greater relative flowability.  
15 The values given in Table I are relative values. The dry flow  
16 of the batch-reacted product made at 360°F. using a  $\text{CaO/SiO}_2$   
17 mol ratio of 0.5 has arbitrarily been assigned a flow number  
18 of "100". A flow number of 200 then indicates that the  
19 powder flowed from the funnel in one-half the time required  
20 for that of the product having a flow number of 100.



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Table I

		Reaction	CaO/SiO <sub>2</sub>	Bulk	Water		X-ray
		Temp.	Mol	Density	Absorption	Dry	Diffraction
		°F	Ratio	lb/cu.	gm/100	Flow-	Identifi-
				ft.	Solids	ability	cation
5	Continuous	360	0.5	13	270	107	CSH-I
6	Batch	360	0.5	7	425	100	CSH-I
7	Continuous	212	0.6	13	260	-	CSH-I
8	Batch	212	0.6	7	470	-	CSH-I
9	Continuous	212	0.3	13	260	-	CSH-I
10	Batch	212	0.3	6	440	-	CSH-I
11	Continuous	450	0.5	20	140	100	CS-55
12	Batch	450	0.5	5	500	56	CS-55
13	Continuous	450	1.0	11	300	76	Xonotlite
14	Batch	450	1.0	4	500	52	Xonotlite

The novel products of this invention as indicated herein-  
before are essentially characterized by their bulk density  
which ranges between approximately 8- and 30 lbs./cu. ft., and  
their Gardner-Coleman Absorption (H<sub>2</sub>O) which preferably ranges  
between 125- and 400-lbs./100-lbs. solids. The products of  
the invention are also characterized by their composite CaO/  
SiO<sub>2</sub> mol ratio between approximately 0.2 and 2.0 to 1, their  
high dry flowability which is believed to be due to the gener-  
ally rounded particle shape. Such product characteristics  
have not been heretofore found in hydrated calcium silicates  
formed by procedures as described in the prior art.

A comparison of the physical appearances and character-  
istics of a batch-reacted hydrated calcium silicate of the  
type described in the prior art, and a continuous reacted  
product of the type defined in this application, may be made  
by reference to Figs. 2 and 3. Fig. 2 is a photomicrograph of

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1 a hydrated calcium silicate formed by a batch-type described  
2 in the afore-mentioned patent to Calvert. Fig. 2 is a photo-  
3 micrograph of a hydrated calcium silicate manufactured on a  
4 continuous basis in accordance with the teaching of this  
5 invention. Fig. 4 is a photomicrograph of a hydrated calcium  
6 silicate manufactured by a batch-process using reaction condi-  
7 tions such that CS-55 was formed. Fig. 5 is a photomicro-  
8 graph of a hydrated calcium silicate manufactured using simi-  
9 lar processing conditions except a continuous process was  
10 used. Process conditions are as described in Example II.  
11 Fig. 6 is a photomicrograph of a hydrated calcium silicate  
12 manufactured by a batch-process using reaction conditions  
13 such that Xonotlite was formed. Fig. 7 is a photomicrograph  
14 of a hydrated calcium silicate manufactured using similar  
15 processing conditions except a continuous process was used.  
16 Process conditions are as described in Example III. More  
17 cursory examination of these photomicrographs immediately  
18 discloses the striking difference of the shape and particle  
19 size characteristics of the materials prepared by batch, and  
20 by continuous processing. The products of this invention  
21 may be seen to have a generally rounded particle structure  
22 which gives them the desirable characteristic of free flow-  
23 ability. The photomicrographs also illustrate the advanta-  
24 geous characteristic of substantial lack of extreme fines in  
25 the product of this invention.

26 A typical example of the application of this invention  
27 is in the conditioning of commercial insecticide dusts. The  
28 product manufactured using the process as described in Example  
29 I can be added in small amounts to the concentrate in order  
30 to improve its dry flowability. For example, 3% of the pro-  
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1 dust produced using reaction conditions described in Example  
2 I was added to a Commercial 40% Chlordane powder, a type of  
3 insecticide described in U. S. Patent No. 2,519,190 and  
4 Insect Control by Chemicals by Brown; Wiley Pub., 1951. The  
5 flowability of the finished dust was improved 20% by the  
6 addition of 3% of the very free-flowing powder. Similarly  
7 the products described in this invention can be added to  
8 other powders to improve their flowability.

9 It is to be appreciated that the products of this  
10 invention have utility for other uses than those described.  
11 For example, the product has been found useful as an insecti-  
12 cide carrier, a catalyst carrier, and an anti-caking agent  
13 for deliquescent salts.  
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SUPPLEMENTARY DISCLOSURE

Whereas the principal disclosure concerns a continuous flow process for the manufacture of hydrated calcium silicates of high bulk density and good absorbency, and products thereof, subsequent investigations have extended the scope and/or resulted in the discovery of a new and inseparable phase or modification of the hereinbefore disclosed invention as well as providing improved and additional unexpected beneficial results.

As pointed out hereinbefore, highly absorbent, bulky hydrated calcium silicates are not suitable for certain applications wherein it is desirable to utilize a hydrothermally prepared hydrated calcium silicate but a product with a relatively low absorption capacity and high bulk density. For example, hydrothermally prepared hydrated calcium silicates are highly effective as dry cleaning sweeteners preventing build up of free fatty acids in dry cleaning solvents. Such hydrated calcium silicate products may be added as a precoat to the dry cleaner's filter and the solvent passed through the precoat where they can be added to the washer and removed at the filter along with the soil. However, a dry cleaner's filter has a limited cake capacity and when the filter cake accumulates to the point of filling the filter chamber it is necessary to stop the operation and remove the cake from the filter. Although calcium silicate products prepared according to the teaching of United States Letters Patent No. 1,574,366 to Calvert are an effective dry cleaning sweetener insofar as acid control, it has the disadvantage of rapidly filling the dry cleaner's filter due to its low cake density. Hence, a hydrated, hydrothermally

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1 prepared, high density calcium silicate having the same  
2 "acid-sorption" per pound would be desirable and advantage-  
3 ous in this particular exemplary application as well as  
4 numerous others.

5 This supplementary portion of the disclosure accordingly  
6 is directed to a more detailed and specific consideration  
7 of the previously disclosed invention of general and specific  
8 phases or modifications thereof. According to additional  
9 investigations relating to this invention, a material and  
10 beneficial bulk density increase and reduction in absorption,  
11 among other advantages, can be achieved in hydrothermally  
12 prepared, particulate hydrated calcium silicate products  
13 by regulating the addition or initial combining of the  
14 reactants. Thus, if a hot suspension of reactive siliceous  
15 material, such as diatomaceous silica, is first fed into a  
16 reaction vessel and the lime suspension subsequently added  
17 thereto, heat being provided to incite reaction and to  
18 provide and maintain the desired reaction conditions, pro-  
19 ducts exhibiting physical as well as chemical properties  
20 comparable to those obtained from a Calvert type reaction,  
21 comprising simply combining reactive siliceous material and  
22 lime in an aqueous medium and then increasing the temperature  
23 of said reactants to reaction temperature, are obtained for  
24 like reaction conditions, i.e., temperature, reaction time,  
25 concentration or liquid to solids content, and reactants.

26 It has been discovered, however, that effecting initial con-  
27 tact of the lime and siliceous reactants by adding or intro-  
28 ducing the siliceous material to a concentrated lime suspen-  
29 sion, for example, first filling the reaction vessel with a  
30 hot lime suspension and subsequently adding the siliceous  
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1 suspension, while maintaining said reactants at a tempera-  
2 ture at least in the vicinity of 250°F. but no less than  
3 212°F. and preferably in the approximate range of 300 to  
4 375°F. results in particulate hydrated calcium silicates of  
5 materially different physical properties such, for example,  
6 as substantially higher bulk densities, lower absorption  
7 characteristics, and larger average particle size. Moreover,  
8 these distinguishing and significant properties are obtained  
9 when identical reactants and reaction conditions are utilized  
10 other than the specified order and temperature of addition  
11 or combining the reactants. Typical property values obtained  
12 from hydrated calcium silicate products prepared according  
13 to a hydrothermal method which included adding the reactive  
14 silicate material to a lime suspension at a temperature of at  
15 least 212°F. and reacting the same for approximately 2 hours  
16 at about 360°F. comprise a fluffed bulk density of about 17  
17 lbs. per cu. ft., and Gardner-Coleman water absorption of  
18 275%.

19 Bulk density increases and water absorption decreases  
20 of even greater magnitude can be obtained in hydrothermally  
21 prepared, particulate hydrated calcium silicates through  
22 initial introduction or addition of the siliceous reactant  
23 into a lime suspension by continuous and simultaneous feeding  
24 of the aqueous suspensions of said siliceous reactant and  
25 lime to a stirred reaction vessel whereby the aqueous suspen-  
26 sion of reactive siliceous material is continuously added or  
27 introduced into an aqueous suspension of lime, retaining the  
28 reactants in said vessel at a temperature and for a reaction  
29 or dwell period sufficient to effect substantial reaction to  
30 result in the particular hydrated calcium silicate desired,  
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1 and continuously and simultaneously with said feeding of the  
2 reactants removing the resulting reaction products from the  
3 reactor at a rate approximately equivalent to the rate of  
4 feed. Typical hydrated calcium silicate product property  
5 values resulting from the foregoing procedure when using a  
6 reaction temperature of approximately 360°F. and a reaction  
7 or retention time of about 2 hours are a fluffed bulk density  
8 of approximately 15 lbs. per cu. ft., a wet cake density of  
9 20 lbs. per cu. ft., and a Gardner-Coleman water absorption  
10 of 240%.

11 The chemical, or physico-chemical mechanism of this  
12 phenomenon is neither fully apparent nor claimed to be com-  
13 pletely understood; however, for purposes of illustration  
14 rather than explanation or limitation, observation indicates  
15 that the physical characteristics of the product are deter-  
16 mined during the period of initial contact and/or reaction  
17 between the reactants, a period when the lime concentration  
18 is at a maximum. It is believed that the reactive nuclei  
19 which form in the initial part of the reaction determine the  
20 physical characteristics of density, particle size and  
21 absorption and it is postulated that the nuclei formed in a  
22 medium of a high lime concentration consist of a calcium rich  
23 phase of calcium silicate. This occurs in a process wherein  
24 the diatomaceous silica is added to a lime suspension and/or  
25 where both reactants are continuously combined or added to a  
26 reaction vessel and new lime suspension is continuously  
27 supplied always providing a high lime concentration during  
28 the initial phase of contact and/or reaction when these  
29 nuclei are formed. On the other hand, the nuclei formed when  
30 the lime is added to the diatomaceous silica suspension  
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1 apparently consist of a calcium silicate with a much lower  
2 calcium content. The nuclei which form in the Calvert type  
3 process consisting of combining the reactants at sub-  
4 reactive temperature and heating to gradually approach and  
5 achieve reactive conditions no doubt also consist of a low  
6 calcium content calcium silicate.

7 The following examples illustrate and compare the prior  
8 art procedures with variations in the practice of the present  
9 invention and the substantial meritorious results thereof  
10 over the prior art practices. It is to be understood that  
11 the specified reactants, reaction conditions such as time,  
12 temperature and concentration, specific techniques or the  
13 like are exemplary and are not to be construed to limit the  
14 invention to the particular components, proportions or other  
15 specified conditions or techniques in the hereinafter des-  
16 cribed examples.

#### 17 EXAMPLE I

##### 18 ADDITION OF LIME TO SILICA SUSPENSION IN A BATCH REACTION

19 Three thousand two hundred and sixty gallons of an  
20 aqueous suspension of diatomaceous earth (silica) comprising  
21 0.92 lbs. of solids per gallon were passed through a heat  
22 exchanger raising the temperature of the same to about 300°F.  
23 and discharged into a pre-heated, stirred reaction vessel  
24 wherein said suspension was further heated to and maintained  
25 at 370°F. by direct injection steam. After addition of the  
26 diatomaceous earth suspension, 1500 gallons of water were  
27 pumped into the reaction vessel over a 40 minute period upon  
28 being similarly pre-heated to a temperature of 300°F. in the  
29 heat exchanger. Simultaneous with the addition of the water,  
30 860 gallons of an aqueous suspension of hydrated lime  
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comprising 1.4 lbs. of CaO per gallon were also added to the reaction vessel, an amount sufficient to result in a CaO/SiO<sub>2</sub> mol ratio of 0.5. Steam was injected into the reactor throughout the addition of said reactants to maintain the temperature of the same at about 370°F. The reaction was continued for a period of 2 hours after all reactants had been added and upon removal from the reactor and filtering of the resulting suspension the cake was dried to a moisture content of about 5% and ground in a hammermill.

#### EXAMPLE II

##### PREMIXING OF LIME AND SILICA AS TAUGHT BY CALVERT IN A BATCH REACTION

An aqueous suspension of hydrated lime, diatomaceous earth (silica) and water was prepared by mixing 1640 lbs. of hydrated lime (1200 lbs. of CaO), 3000 lbs. of diatomaceous silica, and 4500 gallons of water at room temperature to provide a reactive medium of lime and siliceous material having a CaO/SiO<sub>2</sub> mol ratio of 0.5. This suspension was pumped into a cold, stirred reaction vessel and was then heated by direct injection steam to a temperature of 370°F. and maintained at said temperature for a period of 2 hours. Upon completion of the reaction period the resulting suspension was filtered, the cake dried to a moisture content of about 5% and ground in a hammermill.

#### EXAMPLE III

##### ADDITION OF SILICA TO LIME SUSPENSION IN A BATCH REACTION

The reaction conditions in the following example are identical to those of Example I except for the order of addition of the reactants. Eight hundred and sixty gallons of an aqueous suspension of hydrated lime comprising 1.4 lbs.

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1 of CaO per gallon of water were added to a pre-heated,  
2 stirred reaction vessel followed by 1500 gallons of water  
3 and the temperature thereof brought to 370°F. by steam  
4 injection. Next, 3260 gallons (3000 lbs. of diatomaceous  
5 earth) of a slurry of diatomaceous earth (silica) contain-  
6 ing 0.92 lbs. of solids per gallon was pumped through a heat  
7 exchanger bringing its temperature up to 300°F, into the  
8 reaction vessel over an 80 minute period. The ratio of lime  
9 to diatomaceous silica was proportioned to give a  $\text{CaO/SiO}_2$   
10 mol ratio of 0.5. The contents of the reaction vessel were  
11 maintained at 370°F. for 2 hours during which time said con-  
12 tents were stirred by mechanical agitator. Upon completion  
13 of the reaction period the slurry was filtered, the filter  
14 cake dried to a moisture content of about 5% and ground in a  
15 hammermill.

EXAMPLE IVCONTINUOUS ADDITION OF REACTANTS AND CONTINUOUS REMOVAL OF  
THE REACTION PRODUCT

19 An aqueous lime suspension comprising 1.4 lbs. of CaO  
20 per gallon of water and separate aqueous suspension of sili-  
21 ceous material comprising 0.55 lbs. of finely ground dia-  
22 tomaceous silica per gallon of water were continuously and  
23 simultaneously fed into the bottom of the first of 2 stirred  
24 reactors connected in series. The silica suspension being  
25 added at a rate of about 90 gallons per minute and the lime  
26 suspension at a rate of about 16.5 gallons per minute to  
27 provide an approximate  $\text{CaO/SiO}_2$  mol ratio of 0.5. The sep-  
28 arate reactive suspensions were added through separate pipe  
29 lines. The diatomaceous earth suspension was pre-heated in  
30 a heat exchanger to about 300°F, and steam was injected into  
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1 the first reactor to maintain the temperature of its con-  
2 tents at 370°F. When the first reactor reached its overflow  
3 line the suspension of reacting lime and siliceous material  
4 continuously passed over into a second reactor which upon  
5 filling to its overflow line discharged a suspension of the  
6 product continuously and simultaneously at a rate approxi-  
7 mately equivalent to that of the feed. The reacted suspen-  
8 sion was cooled in a heat exchanger and depressurized through  
9 a valve. The reaction time (vessel working capacity divided  
10 by rate of discharge from the second reactor) was about 2  
11 hours; however, several hours are required to reach equilib-  
12 rium in a continuous flow process but once equilibrium is  
13 obtained there was neither an increase nor decrease in the  
14 quantity of material in the reactors since the amount of  
15 reacted suspension removed from the second reactor was equi-  
16 valent to the amount of reactants added to the first reactor.  
17 As in the foregoing examples the suspension from an equilib-  
18 rium condition was filtered, the cake dried and ground in a  
19 hammermill.

20 The solids content of the reacted suspensions, from  
21 each of the foregoing examples were approximately the same:  
22 0.6 to 0.7 lbs. per gallon. The relevant properties of the  
23 ground products of each of the foregoing examples are as  
24 follows:  
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Ex. No.	Fluffed Bulk Density lb./cu. ft.	Wet Cake Density lb./cu. ft.	Gardner-Coleman Water Absorption - %	X-ray Identification
I	7	11	440	Calcium Silicate Hydrate I
II	7	10	430	
III	13	17	260	"
IV	15	20	240	"

Among other unique and beneficial results of the method comprising this invention is the uniformity of the size of the hydrated calcium silicate particles of the reaction product. For example, relative particle size distribution and uniformity of the products of Examples I, III and IV, all consisting of calcium silicate hydrate I materials prepared under identical conditions but for the order of combining the reactants, are as follows:

Size Range	Product of Example I	Product of Example III	Product of Example IV
> 20 microns	10.5%	10.0%	40.0%
10 - 20 microns	19.5	52.0	35.0
8 - 10 microns	10.0	15.0	6.0
6 - 8 microns	15.0	7.0	5.5
4 - 6 microns	22.0	5.5	3.5
2 - 4 microns	12.0	3.0	3.0
< 2 microns	11.0	7.5	7.0

Although certain of the physical properties or characteristics exhibited by the various hydrated calcium silicate products of the method of this invention materially differ from those of comparable hydrated calcium silicate products, i.e., those prepared from like materials, mol ratios, concentrations, reaction time and temperatures, etc., their chemical and other properties such as those established

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1 or determined by X-ray diffraction patterns, thermobalance,  
2 differential thermal analysis remain substantially identi-  
3 cal.

4 Reactants for the practice of the hereinbefore described  
5 invention may comprise finely divided lime and finely divided  
6 reactive siliceous material, such as diatomaceous earth,  
7 quartz, etc., as starting materials. Preferably, finely  
8 divided hydrated lime and diatomaceous silica are employed.  
9 The hydrated calcium silicate products of this invention  
10 may be prepared utilizing these starting materials in a  
11  $\text{CaO/SiO}_2$  mol ratio within the approximate range of 0.1 - 2.0  
12 mole of  $\text{CaO}$  to 1 mol of  $\text{SiO}_2$ . Preferably the slurries or  
13 suspensions of reactive materials include sufficient water  
14 to render their consistency such as to enable it to be  
15 pumped, e.g., approximately 1/2 to 2 lbs. of solids per  
16 gallon of water. Further, the reaction slurry or suspension  
17 should comprise about 8 - 50 parts by weight of water per  
18 part of solids.

19 In accordance with this invention and to effect reaction  
20 between the lime and siliceous components, the reaction  
21 vessel should be maintained at a temperature of at least  
22  $212^\circ\text{F}$ . for a continuous type reaction, and desirably higher,  
23 e.g., in the vicinity of approximately  $250^\circ\text{F}$ . for a batch  
24 type operation to achieve the desired effect. Preferred  
25 reaction temperatures range from about 300 to  $375^\circ\text{F}$ . but  
26 the maximum temperature for the reaction is limited only by  
27 the pressure capacity of the autoclave or vessel and/or the  
28 critical point of water but temperatures within the approxi-  
29 mate range of  $212^\circ\text{F}$ . to  $500^\circ\text{F}$ . can usually be employed.  
30 Further, the  $\text{CaO/SiO}_2$  mol ratio, reaction temperature and  
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1 time, as in the hydrothermal preparation of hydrated calcium  
2 silicates by conventional methods or techniques, each have  
3 an effect on the type of calcium silicate product resulting  
4 from the reaction. For example, relatively low reaction  
5 temperatures, about 370°F. or less, and/or short reaction  
6 periods, result in a compound commonly known as calcium  
7 silicate hydrate I (Taylor, Journal of the Chemical Society,  
8 70, 1953) which can be formed over the entire range of an  
9 0.1 to 2.0 CaO/SiO<sub>2</sub> mol ratio. At a temperature of about  
10 212°F. this reaction, on a continuous basis, has been found  
11 to be substantially complete in approximately 20 minutes.  
12 At relatively higher reaction temperatures, e.g., above  
13 about 370°F., and/or at long reaction periods, and with a  
14 CaO/SiO<sub>2</sub> mol ratio within the range of 0.1 to 0.7, calcium  
15 silicate hydrate I is formed first and then partially or  
16 completely converted to a low solubility hydrated calcium  
17 silicate having the formula 2CaO.3SiO<sub>2</sub>.1~2.5H<sub>2</sub>O and an X-ray  
18 diffraction pattern having very strong lines at  $d = 3.12\text{\AA}$  and  
19  $d = 4.12\text{\AA}$  and a medium line at  $d = 8.34\text{\AA}$ , described in co-  
20 pending Canadian patent application Serial No. 727,526,  
21 filed April 18, 1957. A substantially complete conversion  
22 of calcium silicate hydrate I to the foregoing mentioned  
23 calcium silicate is obtained in about 2 hours when a CaO/SiO<sub>2</sub>  
24 mol ratio of 0.5 to 0.6 is employed in conjunction with a  
25 reaction temperature of 450°F. However, if the CaO/SiO<sub>2</sub> mol  
26 ratio is within the range of 0.8 to 1.5 and the reaction  
27 temperature in excess of about 370°F., calcium silicate  
28 hydrate I also forms first which in turn is partially or  
29 completely transformed to xonotlite. A substantially com-  
30 plete conversion to xonotlite can be obtained in about 2  
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1 hours with a  $\text{CaO/SiO}_2$  mol ratio of 1.0 which is employed  
2 with a reaction temperature of about  $450^\circ\text{F}$ . Reaction times  
3 longer than those necessary to complete the reaction typi-  
4 cally have no detrimental effect upon the physical character-  
5 istics of the resulting products, although, as apparent from  
6 the foregoing, they may result in the conversion of calcium  
7 silicate hydrate I to another hydrated calcium silicate  
8 composition.

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The embodiments of the invention in which an exclusive property or privilege is claimed are defined as follows:

1. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbency which comprises continuously feeding to a reaction vessel finely divided lime and finely divided reactive siliceous material in a  $\text{CaO/SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1 and in an aqueous suspension, maintaining the aqueous suspension of said materials at a temperature of at least approximately  $212^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

2. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbency which comprises continuously feeding to a reaction vessel finely divided lime and finely divided reactive siliceous material in a  $\text{CaO/SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1 and in an aqueous suspension having a water to solids ratio of approximately 4 to 50 parts of water per part of solids, maintaining the aqueous suspension of said materials at a temperature of at least approximately  $212^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

3. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbency which comprises forming aqueous suspensions of finely divided lime and finely divided reactive siliceous material, feeding said

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suspensions to a reaction vessel at a rate sufficient to form an aqueous slurry of lime and reactive siliceous material in a  $\text{CaO/SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1, maintaining the aqueous slurry of said materials at a temperature of at least approximately  $212^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

4. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbcency which comprises forming aqueous suspensions of finely divided lime and finely divided reactive siliceous material, feeding said suspensions to a reaction vessel at a rate sufficient to form an aqueous slurry of lime and reactive siliceous material in a  $\text{CaO/SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1, said slurry having a water to solids ratio of approximately 4 to 50 parts of water per part of solids, maintaining the aqueous slurry of said materials at a temperature of at least approximately  $212^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

5. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbcency which comprises forming aqueous suspensions of finely divided lime and finely divided reactive siliceous material, said suspensions having a solids concentration of approximately 0.1 to 2 lbs. solids per gallon of water, feeding said suspensions to a reaction vessel at a rate sufficient to form an aqueous

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slurry of lime and reactive siliceous material in a  $\text{CaO}/\text{SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1, maintaining the aqueous slurry of said materials at a temperature of at least approximately  $212^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

6. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbency which comprises continuously feeding to a reaction vessel finely divided lime and finely divided reactive diatomaceous earth in a  $\text{CaO}/\text{SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1 and in an aqueous suspension, maintaining the aqueous suspension of said materials at a temperature of at least approximately  $212^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

7. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbency which comprises continuously feeding to a reaction vessel finely divided lime and finely divided reactive diatomaceous earth in a  $\text{CaO}/\text{SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1 and in an aqueous suspension having a water to solids ratio of approximately 4 to 50 parts of water per part of solids, maintaining the aqueous suspension of said materials at a temperature of at least approximately  $212^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

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8. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbency which comprises forming aqueous suspensions of finely divided lime and finely divided reactive diatomaceous earth, feeding said suspensions to a reaction vessel at a rate sufficient to form an aqueous slurry of lime and reactive diatomaceous earth in a  $\text{CaO/SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1, maintaining the aqueous slurry of said materials at a temperature of at least approximately  $212^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

9. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbency which comprises forming aqueous suspensions of finely divided lime and finely divided reactive diatomaceous earth, feeding said suspensions to a reaction vessel at a rate sufficient to form an aqueous slurry of lime and reactive diatomaceous earth in a  $\text{CaO/SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1, said slurry having a water to solids ratio of approximately 4 to 50 parts of water per part of solids, maintaining the aqueous slurry of said materials at a temperature of at least approximately  $212^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

10. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbency which comprises forming aqueous suspensions of finely divided lime

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and finely divided reactive diatomaceous earth, said suspensions having a solids concentration of approximately 0.1 to 2 lbs. solids per gallon of water, feeding said suspensions to a reaction vessel at a rate sufficient to form an aqueous slurry of lime and reactive diatomaceous earth in a  $\text{CaO/SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1, maintaining the aqueous slurry of said materials at a temperature of at least approximately  $212^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

11. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbency which comprises forming aqueous suspensions of finely divided lime and finely divided reactive siliceous material, said suspensions having a solids concentration of approximately 0.1 to 2 lbs. solids per gallon of water, feeding said suspensions to a reaction vessel at a rate sufficient to form an aqueous slurry of lime and reactive siliceous material in a  $\text{CaO/SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1, maintaining the aqueous slurry of said materials at a temperature from approximately  $212$  to  $450^\circ\text{F}$ . for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

12. The method of manufacturing a hydrated calcium silicate having high bulk density and good absorbency which comprises forming aqueous suspensions of finely divided lime and finely divided reactive diatomaceous earth, said suspen-

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sious having a solids concentration of approximately 1/2 to 2 lbs. solids per gallon of water, feeding said suspensions to a reaction vessel at a rate sufficient to form an aqueous slurry of lime and reactive diatomaceous earth in a  $\text{CaO}/\text{SiO}_2$  mol ratio between approximately 0.5 and 1.0 to 1, maintaining the aqueous slurry of said materials at a temperature from approximately 212 to 450°F. for a period of time sufficient to obtain reaction therebetween, and removing the formed reaction product from said vessel at a rate equivalent to the rate of feed of the reactants.

13. A hydrothermally prepared hydrated calcium silicate having a generally rounded, free flowing particle structure and characterized by a bulk density of approximately 8 to 30 lbs./cu. ft. and a Gardner-Coleman Absorption ( $\text{H}_2\text{O}$ ) of approximately 175 to 400 lbs./100 lbs. solids.

14. A hydrothermally prepared hydrated calcium silicate having a generally rounded, free flowing particle structure and characterized by a bulk density of approximately 8 to 30 lbs./cu. ft., a composite  $\text{CaO}/\text{SiO}_2$  mol ratio between approximately 0.1 and 2.0 to 1, and a Gardner-Coleman Absorption ( $\text{H}_2\text{O}$ ) of approximately 175 to 400 lbs./100 lbs. solids.

15. A continuous, hydrothermal method of manufacturing free flowing hydrated calcium silicates having a bulk density of at least approximately 8 lbs./cu. ft. and good absorbcency which comprises continuously and simultaneously feeding finely divided lime and finely divided reactive siliceous material, proportioned to provide a  $\text{CaO}/\text{SiO}_2$  mol ratio of between approximately 0.1 and 2.0 to 1, to a reactor while removing the resulting reaction product from said reactor at a

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rate equivalent to the rate of feed of said reactants, and maintaining an aqueous suspension of said materials within said reactor at a temperature of at least approximately 212°F. for a dwell period sufficient to obtain reaction therebetween.

16. A continuous, hydrothermal method of manufacturing free flowing hydrated calcium silicates having a bulk density of at least approximately 8 lbs./cu. ft. and good absorbency which comprises continuously and simultaneously feeding finely divided lime and finely divided reactive siliceous material, proportioned to provide a  $\text{CaO/SiO}_2$  mol ratio of between approximately 0.1 to 2.0 to 1, in an aqueous suspension having a water to solids ratio of approximately 4 to 50 parts of water per part of solids to a reactor while removing the resulting reaction product from said reactor at a rate equivalent to the rate of feed of said reactants, and maintaining an aqueous suspension of said materials within said reactor at a temperature of at least approximately 212°F. for a dwell period sufficient to obtain reaction therebetween.

17. A continuous, hydrothermal method of manufacturing free flowing hydrated calcium silicates having a bulk density of at least approximately 8 lbs./cu. ft. and good absorbency which comprises continuously and simultaneously feeding finely divided lime and finely divided reactive siliceous material, proportioned to provide a  $\text{CaO/SiO}_2$  mol ratio of between approximately 0.1 and 2.0 to 1, to a reactor while removing the resulting reaction product from said reactor at a rate equivalent to the rate of feed of said reactants, and

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maintaining an aqueous suspension of said materials within said reactor at a temperature of at least approximately 212°F. for a dwell period of at least approximately 20 minutes.

18. A continuous, hydrothermal method of manufacturing free flowing hydrated calcium silicates having a bulk density of at least approximately 8 lbs./cu. ft. and good absorbcency which comprises continuously and simultaneously feeding finely divided lime and finely divided reactive siliceous material, proportioned to provide a  $\text{CaO/SiO}_2$  mol ratio of between approximately 0.1 and 2.0 to 1, in an aqueous suspension having a water to solids ratio of approximately 4 to 50 parts of water per part of solids to a reactor while removing the resulting reaction product from said reactor at a rate equivalent to the rate of feed of said reactants, and maintaining an aqueous suspension of said materials within said reactor at a temperature of at least approximately 212°F. for a dwell period of at least approximately 20 minutes.

19. A continuous, hydrothermal method of manufacturing free flowing hydrated calcium silicates having a bulk density of at least approximately 8 lbs./cu. ft. and good absorbcency which comprises continuously and simultaneously feeding finely divided lime and finely divided reactive siliceous material, proportioned to provide a  $\text{CaO/SiO}_2$  mol ratio of between approximately 0.1 and 2.0 to 1, in an aqueous suspension having a water to solids ratio of approximately 4 to 50 parts of water per part of solids to a reactor while removing the resulting reaction product from said reactor at a rate equivalent to the rate of feed of said reactants, and maintaining an aqueous suspension of said materials within said reactor

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at a temperature of from approximately 212 to 450°F. for a dwell period sufficient to obtain reaction therebetween.

20. A continuous, hydrothermal method of manufacturing free flowing hydrated calcium silicates having a bulk density of at least approximately 8 lbs./cu. ft. and good absorbency which comprises continuously and simultaneously feeding finely divided lime and finely divided reactive siliceous material, proportioned to provide a  $\text{CaO/SiO}_2$  mol ratio of between approximately 0.1 and 2.0 to 1, in an aqueous suspension having a water to solids ratio of approximately 4 to 50 parts of water per part of solids to a reactor while removing the resulting reaction product from said reactor at a rate equivalent to the rate of feed of said reactants, and maintaining an aqueous suspension of said materials within said reactor at a temperature from approximately 212 to 450°F. for a dwell period of at least approximately 20 minutes.

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CLAIMS SUPPORTED BY SUPPLEMENTARY DISCLOSURE

21. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by adding an aqueous suspension of reactive siliceous material to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive siliceous material and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspensions of the siliceous and lime reactants at a temperature of at least 212°F. for a reaction period of at least about 20 minutes to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

22. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. up to approximately 500°F. by adding an aqueous suspension of reactive siliceous material to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive siliceous material and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspension of the siliceous and lime reactants at a temperature of at least 212°F. up to approximately 500°F. for a reaction period

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of at least about 20 minutes to effect substantial reaction between said reactive components to form a hydrated calcium silicate.

23. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature within the range of approximately 300 to 375°F. by adding an aqueous suspension of reactive siliceous material to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive siliceous material and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspension of the siliceous and lime reactants at a temperature within the range of approximately 300 to 375°F. for a reaction period of at least about 20 minutes to effect substantial reaction between said reactive components to form a hydrated calcium silicate.

24. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by adding an aqueous suspension of reactive siliceous material to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive siliceous material and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 - 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids,

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maintaining the aqueous suspensions of the siliceous and lime reactants at a temperature of at least 212°F. for a reaction period of at least about 20 minutes to effect substantial reaction between the reaction components to form a hydrated calcium silicate.

25. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. up to approximately 500°F. by adding an aqueous suspension of reactive siliceous material to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive siliceous material and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 - 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspension of the siliceous and lime reactants at a temperature of at least 212°F. up to approximately 500°F. for a reaction period of at least about 20 minutes to effect substantial reaction between said reactive components to form a hydrated calcium silicate.

26. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature within the range of approximately 300 to 375°F. by adding an aqueous suspension of reactive siliceous material to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive siliceous material and lime to provide a  $\text{CaO/SiO}_2$

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mol ratio of 0.3 - 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspension of the siliceous and lime reactants at a temperature within the range of approximately 300 to 375°F. for a reaction period of at least about 20 minutes to effect substantial reaction between said reactive components to form a hydrated calcium silicate.

27. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by adding an aqueous suspension of reactive siliceous material to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive siliceous material and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspensions of the siliceous and lime reactants at a temperature of at least 212°F. for a reaction period of about 2 hours to effect substantial reaction between said reactive components to form a hydrated calcium silicate.

28. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. up to approximately 500°F. by adding an aqueous suspension of reactive siliceous material to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive

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siliceous material and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 - 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspensions of the siliceous and lime reactants at a temperature of at least  $212^\circ\text{F}$ . up to approximately  $500^\circ\text{F}$ . for a reaction period of about 2 hours to effect substantial reaction between said reactive components to form a hydrated calcium silicate.

29. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature within the range of approximately 300 to  $375^\circ\text{F}$ . by adding an aqueous suspension of reactive siliceous material to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive siliceous material and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 - 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspensions of the siliceous and lime reactants at a temperature within the range of approximately 300 to  $375^\circ\text{F}$ . for a reaction period of about 2 hours to effect substantial reaction between said reactive components to form a hydrated calcium silicate.

30. A method of hydrothermally preparing hydrated calcium silicates having bulk densities of at least approximately 10 lbs. per cu. ft. which comprises effecting initial contact of siliceous and lime reactants at a temperature within the range of approximately 300 to  $375^\circ\text{F}$ . by adding an aqueous suspension of reactive siliceous material to an aqueous

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suspension of lime, proportioning the reactants comprising water and said suspensions of reactive siliceous material and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 - 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspensions of the siliceous and lime reactants at a temperature within the range of approximately 300 to 375°F. for a reaction period of about 2 hours to effect substantial reaction between said reactive components to form a hydrated calcium silicate.

31. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by continuous addition of an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous feeding of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the reactive contents of said vessel at a temperature of at least 212°F, continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of feed, and retaining the aqueous suspension of

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said reactants within said vessel for a reaction period of at least about 20 minutes to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

32. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. up to approximately 500°F. by continuous addition of an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous feeding of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the reactive contents of said vessel at a temperature of at least 212°F. up to approximately 500°F., continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of feed, and retaining the aqueous suspension of said reactants within said vessel for a reaction period of at least about 20 minutes to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

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33. A method of increasing the bulk density of hydro-thermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature within the range of approximately 300 to 375°F. by continuous addition of an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous feeding of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the reactive contents of said vessel at a temperature within the range of approximately 300 to 375°F. continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of feed, and retaining the aqueous suspension of said reactants within said vessel for a reaction period of at least about 20 minutes to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

34. A method of increasing the bulk density of hydro-thermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by continuous addition of an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous

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feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous feeding of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 - 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the reactive contents of said vessel at a temperature of at least  $212^\circ\text{F}$ ., continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of feed, and retaining the aqueous suspension of said reactants within said vessel for a reaction period of at least about 20 minutes to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

35. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least  $212^\circ\text{F}$ . up to approximately  $500^\circ\text{F}$ . by continuous addition of an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous feeding of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 - 1.5 and a reaction medium having a water to solids

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content of about 8 - 50 parts by weight of water per part of solids, maintaining the reactive contents of said vessel at a temperature of at least 212°F. up to approximately 500°F. continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of feed, and retaining the aqueous suspension of said reactants within said vessel for a reaction period of at least about 20 minutes to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

36. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature within the range of approximately 300 to 375°F. by continuous addition to an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous feeding of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 - 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids; maintaining the reactive contents of said vessel at a temperature within the range of approximately 300 to 375°F., continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of

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feed, and retaining the aqueous suspension of said reactants within said vessel for a reaction period of at least about 20 minutes to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

37. A method of increasing the bulk density of hydrothermally prepared hydrated calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by continuous addition of an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous feeding of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the reactive contents of said vessel at a temperature of at least 212°F., continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of feed, and retaining the aqueous suspension of said reactants within said vessel for a reaction period of about 2 hours to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

38. A method of increasing the bulk density of hydrothermally prepared calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. up to approximately 500°F. by

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continuous addition of an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous feeding of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 to 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the reactive contents of said vessel at a temperature of at least  $212^\circ\text{F}$ . up to approximately  $500^\circ\text{F}$ ., continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of feed, and retaining the aqueous suspension of said reactants within said vessel for a period of about 2 hours to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

39. A method of increasing the bulk density of hydrothermally prepared calcium silicates which comprises effecting initial contact of siliceous and lime reactants at a temperature within the range of approximately 300 to  $375^\circ\text{F}$ . by continuous addition of an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous feeding

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of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 to 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the reactive contents of said vessel at a temperature within the range of 300 to 375°F., continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of feed, and retaining the aqueous suspension of said reactants within said vessel for a period of about 2 hours to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

20. A method of hydrothermally preparing hydrated calcium silicates having a bulk density of at least approximately 10 lbs. per cu. ft. which comprises effecting initial contact of siliceous and lime reactants at a temperature within the range of approximately 300 to 375°F. by continuous addition of an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous feeding of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.3 to 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the reactive contents of said vessel at a temperature within the range of

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300 to 375°F., continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of feed, and retaining the aqueous suspension of said reactants within said vessel for a period of about 2 hours to effect substantial reaction between the reactive components to form a hydrated calcium silicate.

41. A hydrothermal method of preparing a calcium silicate hydrate I product with increased bulk density which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by adding an aqueous suspension of reactive diatomaceous earth to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive diatomaceous earth and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspension of diatomaceous earth and lime reactants at a temperature of 212 to 370°F. for a reaction period of at least about 20 minutes up to approximately 24 hours to effect substantial reaction between the reactive components to form calcium silicate hydrate I.

42. A hydrothermal method of preparing a hydrated calcium silicate product having the formula  $2\text{CaO} \cdot 3\text{SiO}_2 \cdot 1-2.5\text{H}_2\text{O}$  with increased bulk density which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by adding an aqueous suspension of reactive diatomaceous earth to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions

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of reactive diatomaceous earth and lime to provide a CaO/SiO<sub>2</sub> mol ratio of 0.1 - 0.7 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspension of diatomaceous earth and lime reactants at a temperature of at least about 370°F. for a reaction period of at least about 20 minutes up to approximately 24 hours to effect substantial reaction between the reactive components to form a hydrated calcium silicate having the formula  $2\text{CaO} \cdot 3\text{SiO}_2 \cdot 1-2.5\text{H}_2\text{O}$ .

43. A hydrothermal method of preparing the hydrated calcium silicate xonotlite with increased bulk density which comprises effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by adding an aqueous suspension of reactive diatomaceous earth to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive diatomaceous earth and lime to provide a CaO/SiO<sub>2</sub> mol ratio of 0.8 to 1.5 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspension of the diatomaceous earth and lime reactants at a temperature of at least 370°F. for a reaction period of at least about 20 minutes up to approximately 24 hours to effect substantial reaction between the reactive components to form xonotlite.

44. A hydrated calcium silicate product comprising discrete particles at least 50% of which are greater than 8 microns and having a bulk density of at least 10 lbs. per cu. ft. and wet cake density of at least 14 lbs. per cu. ft.,

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said calcium silicate consisting of the hydrothermal reaction product of effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by adding an aqueous suspension of reactive siliceous material to an aqueous suspension of lime, proportioning the reactants comprising water and said suspensions of reactive siliceous material and lime to provide a  $\text{CaO/SiO}_2$  mol ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by weight of water per part of solids, maintaining the aqueous suspensions of the siliceous and lime reactants at a temperature of at least 212°F. for a reaction period of at least about 20 minutes to effect substantial reaction between the components.

45. A hydrated calcium silicate product comprising discrete particles at least 50% of which are greater than 8 microns and having a bulk density of at least 12 lbs. per cu. ft. and a wet cake density of at least 16 lbs. per cu. ft., said calcium silicate consisting of the hydrothermal reaction product of effecting initial contact of siliceous and lime reactants at a temperature of at least 212°F. by continuous addition of an aqueous suspension of reactive siliceous material to an aqueous suspension of lime through continuous and simultaneous feeding of the siliceous and lime suspensions to a heated reaction vessel to maintain an uninterrupted introduction of said siliceous reactant into the said lime reactant, proportioning the continuous and simultaneous feeding of the reactants comprising water, siliceous material and lime to the reaction vessel to provide a  $\text{CaO/SiO}_2$  mol. ratio of 0.1 - 2.0 and a reaction medium having a water to solids content of about 8 - 50 parts by

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weight of water per part of solids, maintaining the reactive contents of said vessel at a temperature of at least 212°F., continuously and simultaneously with said feeding of the reactants removing the resulting reaction products from said reactor at a rate approximately equivalent to the rate of feed, and retaining the aqueous suspension of said reactants within said vessel for a reaction period of at least about 20 minutes to effect substantial reaction between the reactive components.